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Annealing of Co_xCu_{1-x} / Cu Multilayers

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ABSTRACT

In the multilayer system cobalt / copper at the second antiferromagnetic coupling maximum (2. AFM) with a copper thickness of d_{Cu} = 2,2 nm it is possible to reduce magnetoresistive hysteresis by the use of either very thin Co-layers or by alloyed magnetic layers $Co_{1-x}Cu_x$. It was possible to achieve values for the giant magnetoresistance effect of GMR ≈ 20 % for as prepared samples. A heat treatment was applied to study the degeneration of the system. For annealing at moderate temperatures ($T_{anneal} \leq 250°C$) an increase up to GMR ≈ 25 % was observed. Annealing at slightly higher temperatures lead to an rapid decrease in the GMR effect. To study the structural changes the method of x-ray reflectivity was utilized showing changes in interface roughness as well as in bilayer thickness.

INTRODUCTION

Since the discovery of the Giant Magnetoresistance effect (GMR)[1] there has been great effort to utilize it for industrial applications. The first branch that used GMR based devices was the computer industry. The assemblies are called Spin-Valves and are used in read heads of hard disc drives. In principle these devices consist of two ferromagnetic films with different coercivities separated by a nonmagnetic material. The thickness of the nonmagnetic layer must be high enough so that there is no magnetic interaction between the ferromagnetic layers.

Another basic principle is the use antiferromagnetically coupled multilayers based on Co and Cu. To reduce undesirable hysteresis Co is often alloyed with Fe[2]. Some alternative approaches are discussed in literature like reducing the thickness of the magnetic layer[3] or alloying the magnetic layer with the material of the spacer layer[4].

Besides the reduction of the hysteresis the main duty is to guaranty the capability of the functional layers to withstand the high temperatures of complete production process of a magnetic sensor ($T_{standard} \approx 300°C$)[2]. The equilibrium phase diagram[5] of Co and Cu shows that the two elements are not miscible promising a relative stable layer configuration with no intermixing. Nevertheless the preparation in UHV by sputtering with high energies may lead to implantation of the atoms of one species into the layer of the other element. As the dimensions of the single layers are in the nanoscale and the GMR is known to be an interfacial effect the control of the interface structure and the knowledge of its changes due to thermal annealing is of great importance for the successful application of GMR devices. There are different approaches to obtain the desired flat interfaces. One is the use of so called buffer layers made of Fe, $Ni_{80}Fe_{20}$, or Ta[6]. The other is the utilization of surfactants like Pb or Ag[7], which float on the surface during the preparation process and inhibit island like growth leading to ferromagnetic pinholes[8]. To enhance the thermal stability it is suggested to alloy the spacer layer with a partially miscible element. This should lead to the formation of an alloy at the grain boundary region of the crystallites of the spacer layer, which should avoid the grain growth connected to thermal annealing. A method combining surfactant mediated growth and formation of stabilizing alloys is the use of targets

consisting of Cu, Ag, and Au[9]. Ag is not miscible with the other components and has a high surface tension, so it floats on the surface through the whole deposition process. Au is partially miscible with Cu and forms the desired interface alloy inhibiting the grain growth.
This study is focused on the annealing behavior of the basic Co / Cu system with alloyed magnetic layer $Co_{1-x}Cu_x$ and varying thickness d_{Co}. The magnetoresistive properties are measured by the standard four-point probe, the structural development is investigated by x-ray reflectivity-techniques.

EXPERIMENTAL DETAILS

Preparation

The $Co_{1-x}Cu_x$ / Cu multilayers were prepared by the use of a commercial magnetron sputtering facility having three positions for sputter cathodes. The variation of the Cu content in the magnetic layer was possible due to a triple configuration on one of the cathode-places. One of these triple positions was equipped with a target of pure Cu another with pure Co. The Cu spacer layer was prepared by an independently working cathode with another pure Cu target. The film thickness was controlled by the sputtering power and the residence time underneath each cathode. The substrate was a Si wafer with 850 nm of thermally oxidized SiO_2. In preliminary experiment the copper thickness d_{Cu} was varied to obtain the desired thickness for the 2nd AFM. It was found to be $d_{Cu} = 2,2$ nm. For defined growth conditions with flat interfaces a $Ni_{80}Fe_{20}$ buffer layer with $d_{buf} = 4$ nm was first deposited on the substrate. The base pressure was better than $5*10^{-7}$ mbar. The sputtering gas was Ar of a pressure of $4*10^{-3}$ mbar. The substrate temperature was 20 °C.
Two sets of samples have been prepared:

i) alloyed magnetic layer, $d_{CoCu} = 1,5$ nm, $0 \leq x_{Cu} \leq 0,48$
ii) pure magnetic layer, $0,5 \text{ nm} \leq d_{Co} \leq 1,5$ nm

The thickness of the spacer layer was $d_{Cu} = 2,2$ nm for all samples corresponding to the 2nd AFM. The number of bilayers was N = 24.

Annealing

The annealing was preformed in a vacuum furnace with a pressure of $p \leq 1*10^{-7}$ mbar with a annealing time of $t_{anneal} = 1$ h in the temperature range of $150°C \leq T_{anneal} \leq 500°C$ in subsequent annealing steps. The heating- and cooling rates were 10 K/min.

Characterization

The measurement of the GMR curves was done by a standard four-point probe at room temperature as well on the as prepared samples as on the samples after each annealing step. The sensing current was flowing in the film plane (CIP geometry) and was parallel to the external magnetic field created by an electromagnet and measured by a Hall probe. For a complete recording of all significant values the field was first set to a maximum value of + 300 mT and was lowered stepwise to the same negative value of – 300 mT. From there the same stepwise measurement was carried out to the starting point. So it was possible to determine the magnetic hysteresis exactly.
X-ray reflectivity measurements were applied for the characterization of the film thickness, interface roughness and periodicity of the multilayered system. The apparatus (Siemens D5000) was equipped with a Cu tube and a secondary monochromator. The incident angle was varied

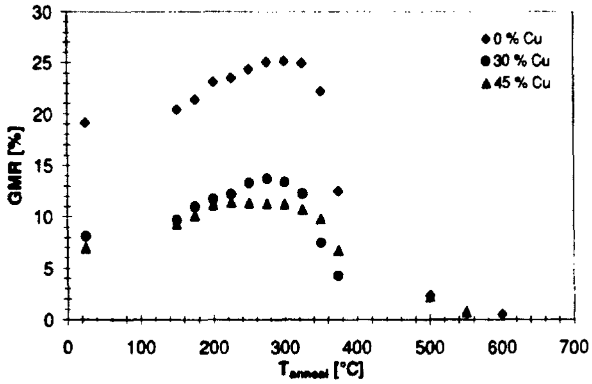

Figure 1. The GMR effect as a function of annealing temperature T_{anneal} for the system with d_{CoCu} = 1,5nm and d_{Cu} = 2,2 nm

from 0,2° ≤ θ ≤ 3°. A part of the X-rays is reflected at the sample surface, another part at inner surfaces. Interference between these reflected parts cause a characteristic oscillation of the intensity depending on the incident angle θ (Kiessig-fringes). The measured reflctograms can be approximated by simulated ones. The parameters of the simulations are the electron density, the thickness of the single layers and the interface roughness.
The superimposed bilayer periodicity causes additional interferences (Bragg-peaks). The position θ_{Bragg} allows the calculation of the bilayer thickness.

DISCUSSION

Alloying of the magnetic layers yielded to a lower GMR effect compared unalloyed system (see Figure 1). Remarkable is the big improvement of 4 % to 5 % for all systems due to the soft annealing. The maximum GMR effect was observed for T_{anneal} ≈ 250°C – 275°C. Annealing at higher temperatures (T_{anneal} ≥ 300°C) caused a rapid decrease in GMR. This limiting temperature was almost the same for all Cu concentrations.
The dependence of the hysteresis on the annealing temperature and the Cu concentration is shown in Figure 2. Alloying of the magnetic layer with 30 % of Cu leads to a reduction of the hysteresis from 6 mT to 1 mT, which is within the specifications. Annealing at temperatures that give rise to the increase of the GMR cause no significant change in the hysteresis. If the threshold of the rapid decrease is exceeded the hysteresis is increasing. This increase is more distinct for the unalloyed samples than for the alloyed one.
The results for the unalloyed system with varying d_{Co} are presented in Figure 3 and Figure 4. The decrease in GMR due to reduction of the thickness of the magnetic material is clearly visible. Even for the thinnest layer with d_{Co} = 0,56 nm the GMR is still ≈ 10 % whereas alloying lead to further reduction (GMR_{min} ≈ 7 % for x_{Cu} = 45 %) . The lowering of the hysteresis is not as significant as in the alloyed case.

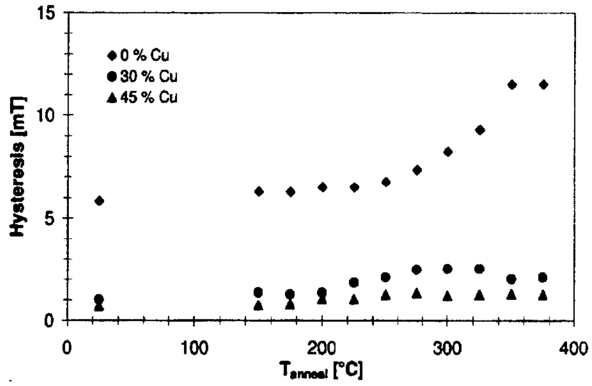

Figure 2. Dependence of the hysteresis on T_{anneal} for different Cu-concentrations in the magnetic layer of the thickness $d_{CoCu} = 1,5$ nm ($d_{Cu} = 2,2$ nm)

The annealed unalloyed samples show a similar rapid degradation for higher annealing temperatures. The threshold temperature of this degeneration is depending on the thickness of the magnetic layer d_{Co}. The thinner the magnetic layer the lower is the limiting temperature. An improvement due to soft annealing is also found but is limited to $d_{Co} \geq 1,02$ nm. The improve is more pronounced for the sample with the thickest magnetic layer. If $d_{Co} < 1,0$ nm there is no improve in GMR. The dependence of the hysteresis on the thickness of the magnetic layer is comparable to that of the GMR effect. For $d_{Co} \geq 1,02$ nm a drastic increase is observed if a threshold temperature of $T_{anneal} \approx 275°C$.

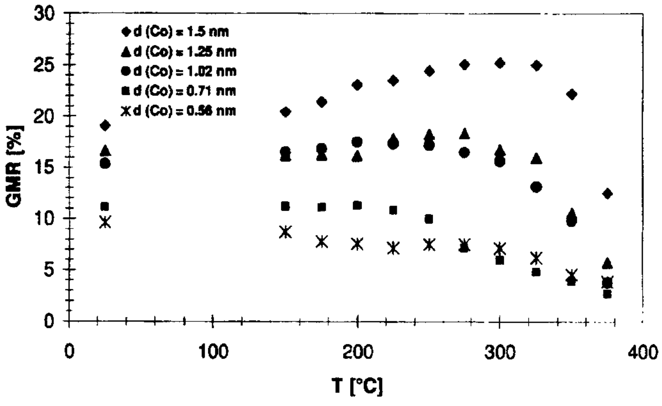

Figure 3. Dependence of the GMR effect on the annealing temperature T_{anneal} and the thickness of the magnetic layer d_{Co} ($d_{Cu} = 2,2$ nm)

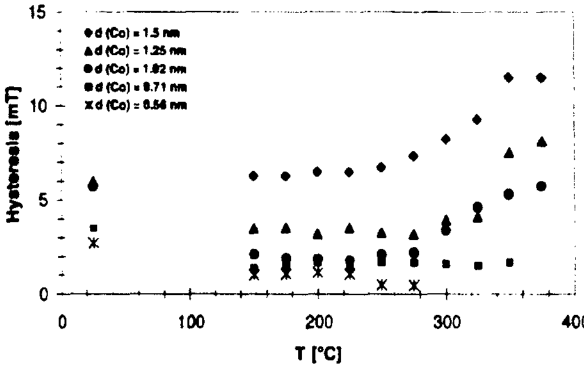

Figure 4. Dependence of the hysteresis on the annealing temperature T_{anneal} and the thickness of the magnetic layer d_{Co} (d_{Cu} = 2,2 nm)

The hysteresis of samples with thinner magnetic layers is relatively stable.
Since the GMR effect is known to be an interface phenomenon the interface sensitive method of X-ray reflectivity measurements has been applied after selected stages of the heat treatment. A sequence of the obtained curves is shown in Figure 5 for the unalloyed systems with d_{Co} = 1,5 nm. The reflectogram of the as prepared sample is compared to the just before the beginning of the rapid decrease and to a sample which showed no GMR at all after though heat treatment at T_{anneal} = 600°C for t_{anneal} = 8 h. The X-ray reflectivity experiments reveal an increase in bilayer thickness due to the soft annealing (shift of the Bragg-peak to smaller angels). The increased intensity of the Bragg peak gives rise to the assumption that the interfaces between Co and Cu have been sharpened.

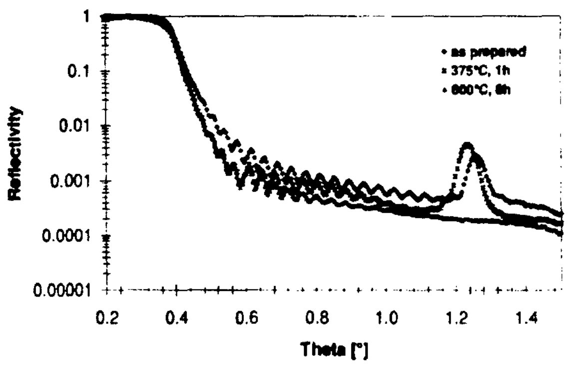

Figure 5. X-ray reflectivity curves for different stages of the heat treatment of unalloyed samples with d_{Co} = 1,5 nm.

The geometrical roughness has increased as indicated be the steeper decay of the curve and the less pronounced Kiessig fringes. The toughly treated sample shows no Bragg-peak any more meaning the total destruction of the bilayer structure.

CONLUSIONS

Based on these results and unpublished data[10, 11] a model for the development of the microstructure and its influence on the GMR effect is developed. In as prepared samples the interfaces are characterized by an intermixing of Co and Cu due to the deposition process. This causes a non-optimal antiferromagnetic coupling and a comparable high basic resistance. The soft annealing initiates out-diffusion of the atomic species at the interfaces. This process lowers the basic resistance and improves the antiferromagnetic coupling and thus the GMR effect. This phenomenon occurs although the geometrical roughness of the interfaces increases. Annealing above a critical temperature results in the further increase of the interface roughness and the onset of discontinuities of the interfaces leading to a decrease of the magnetoresistance.
It is possible to reduce the hysteresis by alloying Co with Cu or by the use of thin Co-layers. Especially for thinner magnetic layers a reduction of the critical temperature for the onset of the degeneration has been observed. The materials selection always has to be a compromise between high GMR, low hysteresis, and desired thermal stability.

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